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(71) Applicant  
The Secretary of State for Defence  
Whitehall, London, SW1A 2HB, United Kingdom

(72) Inventors  
David Victor Dunford  
Peter George Partridge

(74) Agent and/or Address for Service  
S R Skelton  
Procurement Executive, Ministry of Defence,  
Patents 1/A/4, Room 2014, Empress State Building,  
Little Road, London, SW6 1TR, United Kingdom

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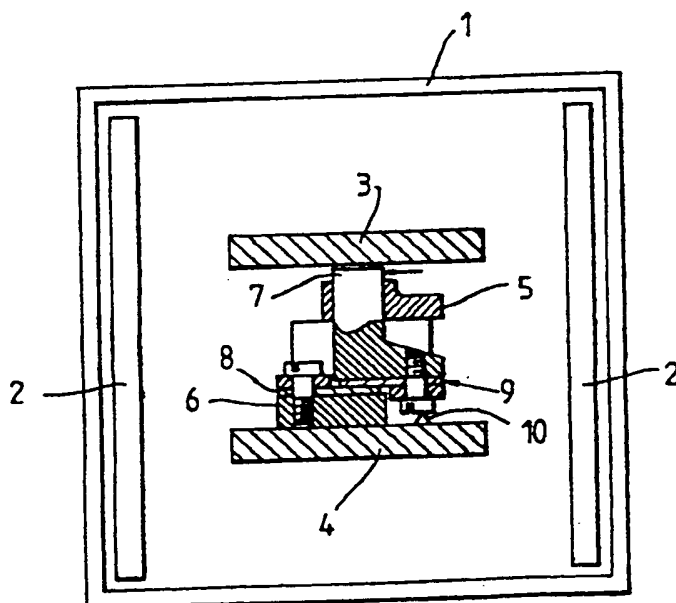
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(54) Diffusion bonding of aluminium-lithium alloys

(57) Pieces of aluminium-lithium alloy including reinforced composites thereof are diffusion bonded without prior surface coating by a process comprising initial mechanical removal of surface oxide and lithium depleted material, smoothing the faying surfaces, placing the pieces whilst space apart into a vacuum chamber, evacuating same, pressing the faying surfaces heavily together, raising their temperature whilst decreasing the applied pressure, and holding the pieces for at least one hour at a temperature in the range 500 to 580°C whilst under light contact pressure. The freedom of movement of lithium within the joint pieces and its ability to disrupt the reoxidised surface layer circumvents any necessity for prevention of reoxidisation when a suitable diffusion bonding regime is utilised for the bonding process.

Fig.1.



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Fig.1.

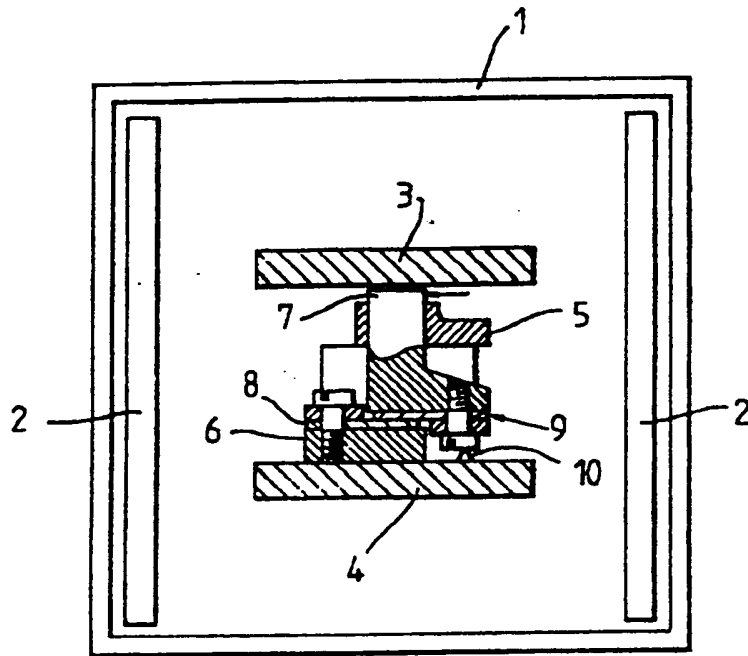
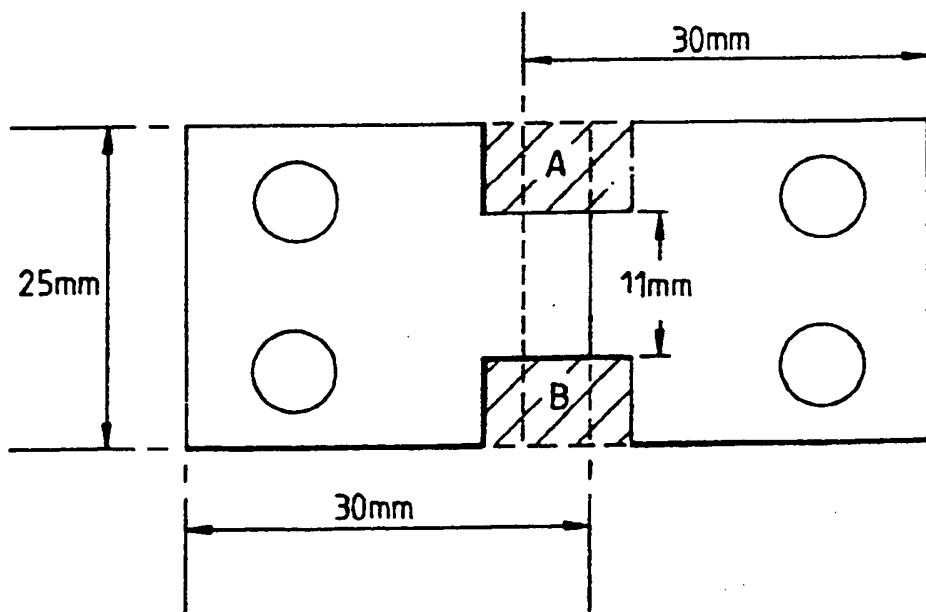


Fig.2.



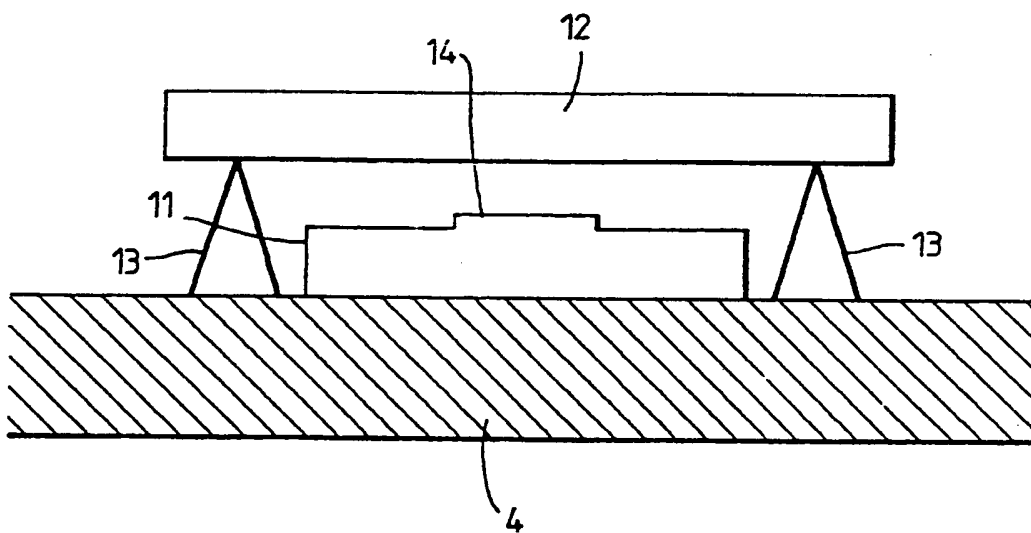
*Fig. 3.*

Fig. 4.

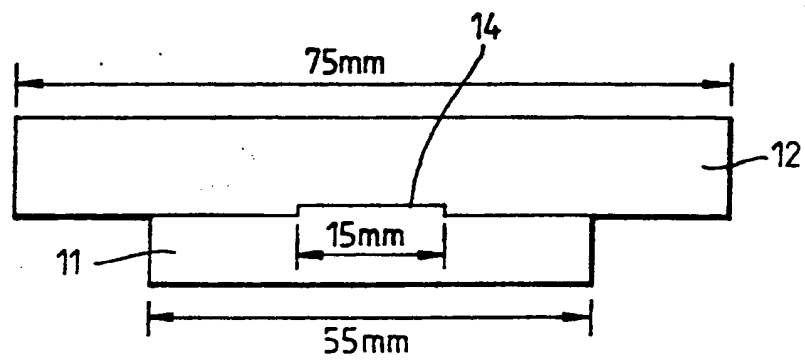
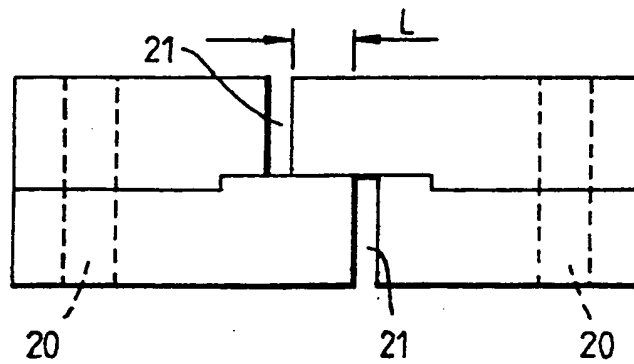


Fig. 5.



## DIFFUSION BONDING PROCESS FOR ALUMINIUM-LITHIUM ALLOYS

This invention relates to a diffusion bonding process by which aluminium-lithium alloy pieces may be bonded to each other. The process is suitable both for joints between unreinforced aluminium-lithium alloys and for joints in which one or each of the pieces to be joined is a metal matrix composite comprising an aluminium-lithium alloy matrix reinforced with ceramic whiskers or particles. In this specification and claims the term "aluminium-lithium alloy" is used to mean such alloys having aluminium as principal ingredient which have at least one percent by weight of lithium.

Aluminium-lithium alloys are attractive materials for use in aircraft structures and the like in place of other established high strength aluminium alloys because of their lower density and increased stiffness. Present day commercial alloys of the aluminium-lithium system are made by conventional casting route techniques rather than rapid solidification techniques and they typically have between 2 and 3% by weight of lithium, with additional strengthening ingredients such as copper and magnesium. Diffusion bonding is a well known bonding method by which metal components may be bonded under the application of heat and pressure by solid state diffusion of atoms across the faying surfaces possibly aided by the creation of a transient liquid phase at the interface. It has long been recognised that it is difficult to produce good diffusion bonds between articles consisting of the established high strength aluminium alloys which do not contain lithium. This difficulty stems from the affinity of aluminium for oxygen and the barrier to atomic diffusion presented by

oxides of aluminium.

Many aluminium alloys exhibit superplastic forming properties and it would be desirable to be able to make aluminium alloy aircraft components by the diffusion bonding/superplastic forming process (DBSPF) as used today for titanium materials. Hitherto the adoption of such a process for aluminium alloy materials has been held back by the difficulty in securing adequate diffusion bonds in such materials.

It is known that aluminium-lithium alloys may exhibit superplastic forming properties but there has been no reason to suppose that these alloys would be any easier to diffusion bond than other aluminium alloys. Indeed it has been reported previously that aluminium-lithium alloys oxidize more readily than other aluminium alloys and reported elsewhere that they are more difficult to diffusion bond than the other aluminium alloys.

A typical diffusion bonding process as used hitherto for non lithium-containing aluminium alloys might embrace ion bombardment cleaning at low pressure followed immediately by ion plating with a metal such as silver or copper to prevent reoxidation before heating and pressing in a regime intended to ensure migration of the plating ingredient from the joint region.

It has now been discovered by those named as inventors for this invention, that aluminium-lithium alloys can be diffusion bonded more easily than other high strength aluminium alloys in regard to the precautions and procedures that need to be followed in order to create joint surfaces suitable for bonding. The lithium within an aluminium-lithium alloy tends to diffuse to the surface of the material when at elevated temperatures - such as the temperatures used in hot working or even diffusion bonding of the material - despite the inevitable surface skin of aluminium oxide. Lithium diffusing to the surface is oxidised but unlike the aluminium oxide layer it is not a stable self healing film. This is manifest as a lithium depleted layer in hot worked products. Passage of the lithium through the aluminium oxide layer tends to disrupt and break up this layer. This is beneficial in the context of a diffusion bonding process for that

aluminium oxide layer would otherwise act as a diffusion barrier prejudicial to successful bonding. The aforementioned lithium depleted layer, in an alloy containing magnesium to a significant degree, is likely to be denuded of magnesium also by the same sort of mechanism but to a lesser degree.

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The inventors have discovered that it is possible to diffusion bond aluminium-lithium alloys successfully by a process making use of the characteristics of the lithium providing any zone of lithium depletion is removed from the joint surfaces before they are bonded. In this process no plating of the prepared joint surfaces for prevention of reoxidation is required as any reformed oxide layer is disrupted to a sufficient extent in bonding by the through-diffusion of lithium.

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The claimed invention is a process by which two or more aluminium-lithium alloy pieces are bonded together by the following steps in sequence.

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machining or abrading material from each faying surface to a depth sufficient to remove existing surface oxide and the sub-surface zone of lithium-depleted material where such exists;

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grinding or polishing the faying surfaces to provide a smoothed surfaces for mating and cleaning these surfaces of polishing residues;

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locating the pieces in a vacuum chamber and evacuating the chamber whilst the faying surfaces are either kept apart or under light contact pressure to allow desorption of surface contaminants;

pressing the pieces together heavily at the faying surfaces whilst maintaining the vacuum and increasing the temperature of the pieces in a regime in which the pressure is eased as the temperature is raised; and;

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holding the pieces together under light pressure for at least one hour at a temperature in the range of 500 to 580°C.

Preferably the initial heavy pressure is in the range 100-120 MPa and preferably also the subsequent light pressure is in the range 0.5-3 MPa.

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The process as thus defined can be applied without variation in

joining one piece of unreinforced alloy to another, in joining one piece of unreinforced alloy to a piece of reinforced alloy and in joining one piece of reinforced alloy to another. However reinforced alloys are much harder than unreinforced alloys and it might be difficult to secure a satisfactory degree of contact in joining one piece of same to another or to an unreinforced alloy. So it may be desirable in such circumstances to interpose a metal film of unreinforced alloy having the same or similar composition to the adjacent materials in order to achieve a sufficient degree of intimacy of contact to permit diffusion bonding at a reduced level of pressure to that which would otherwise be required. The foil interlayer does not need to be machined or surface polished in the manner of the faying surfaces. However, the foil interlayer should preferably be held apart from the faying surfaces until the chamber has been evacuated so that it does not interfere with the evacuation of desorped surface contaminants.

In a further variation of the method, a piece of a dissimilar metal foil, such as a foil of copper or silver, may be used as an interlayer instead of a foil of similar metal in order to promote a bonding regime employing a transient liquid phase.

The diffusion bonding process claimed is described below by way of example with reference to the drawings, of which:

Figure 1 is a cross sectional view of a vacuum hot press with a bonding jig therein;

Figure 2 is a plan view of a lap joint shear stress test piece, produced using the apparatus shown in Figure 1;

Figure 3 illustrates a layout of pieces for diffusion bonding, for the production of a second form of shear stress test piece;

Figure 4 is a sectional view of the bonded pieces; and

Figure 5 is a sectional view of this second form of shear stress test piece.

All the bonding joints described here are made between unreinforced pieces of 8090 alloy sheet in either 2.5mm or 4.0mm gauge except for those joints specifically stated to utilize 8090 alloy in a silicon carbide grit reinforced metal matrix composite. 8090 alloy



has a nominal composition by weight as follows: Al - 2.5% Li - 1.3% Cu - 0.8% Mg - 0.12% Zr - 0.1% maximum Fe - 0.05% maximum Si.

5 The alloy sheet is first cut to the required configuration of the test piece blanks and then the faying surfaces or whole surface of the blanks was ground using a rotary grinder to remove at least 40  $\mu\text{m}$  depth of material. It has been found to be essential to the success of the joining process that not only is the layer of existing aluminium oxide removed but that also the bulk of any layer of lithium depletion is removed. The oxide layer will reform but this in itself is not harmful providing there is no extended delay between the grinding operation and the subsequent bonding and provided also that there is the full measure of lithium at the surface.

10 After grinding the pieces to remove the zone of lithium depleted material the faying surfaces are smoothed for mating by polishing with 1200 grade silicon carbide grit or preferably with 1  $\mu\text{m}$  diamond powder. This provides a good surface contact between faying surfaces. Potential residues from the polishing stage are removed by solvent cleaning in acetone. The cleaning fluid is then allowed to evaporate before the bonding stage and contact with water is avoided.

20 At this stage the pieces are ready to be bonded and are placed within a vacuum hot press. Some form of bonding jig might be required in order to hold the pieces in the required arrangement. Figure 1 shows a typical vacuum hot press comprising an enclosure 1 heating elements 2 and press platens 3 and 4. A bonding jig 5 of a form suitable for preparing lap joint shear stress test pieces is depicted between the press platens 3 and 4. The jig 5 is made in two parts (6 and 7) one being slidably mounted within the other and the metal pieces to be joined (8 and 9) are each bolted to a respective part of the jig so that they are maintained in parallel alignment with a predetermined degree of joint overlap when pressed together for bonding. Until the vacuum chamber has been evacuated to the required degree the two pieces to be bonded are held apart from one another, to permit desorption of surface contaminants, by wedging apart the two parts of the bonding jig 5 by means of a collapsable spacer formed by a folded strip of aluminium foil 10.

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The press enclosure 1 is then evacuated whilst being left at room temperature until the pressure reduced to  $2.7 \times 10^{-5}$  Pa. Thereafter this vacuum pressure level is maintained for the bonding cycle whilst the press platens 3 and 4 were pressed together and the press enclosure 1 is heated by heaters 2. The temperature of the bonding pieces is determined by means of a thermocouple attached to one of them. The pressure/temperature schedule of the bonding cycle is as follows:-

1. apply initial platten pressure of 120 MPa and hold;
2. heat enclosure until bonding pieces reach  $250^{\circ}\text{C}$ , this being attained in approximately 11 minutes;
3. reduce platten pressure to 7 MPa and hold;
4. heat enclosure until bonding pieces reach  $500^{\circ}\text{C}$ , this being attained in approximately 25 minutes;
5. reduce platten pressure to a predetermined level in the band 0.75-1.5 MPa and hold;
6. heat enclosure until bonding pieces obtain  $560^{\circ}\text{C}$ , this being attained in approximately 12 minutes; and
7. hold bonding pieces at  $560^{\circ}\text{C}$  under pressure at the same predetermined level for a required period of between 1 and 19 hours.

Thereafter the bonded pieces are cooled and released for further processing. Simple overlap diffusion bonded test pieces are produced in this way from 25mm x 30mm bonding pieces. After bonding as described and before testing the test pieces are recovery heat treated to the solution treated and aged condition. The solution treatment comprises a heating to  $530^{\circ}\text{C}$ , a hold at temperature for 20 minutes and air or water quenching. The ageing treatment comprises holding the material for 5 hours at  $185^{\circ}\text{C}$  followed by air cooling. Test pieces to the configuration shown in Figure 2, which are called herein "type 1" test pieces, are produced from the 25mm x 30mm bonding pieces, bonded with a required overlap length  $\ell$  by removing sections A and B of the joint so as to leave a nominal joint width of 11mm. Section A is removed after bonding but before the recovery heat treatment and section B is removed after the recovery heat treatment. Removal of these sections eliminates test result scatter caused by edge effects

and provides convenient specimens for examination of joint microstructure in the two metallurgical conditions. The degree of deformation of section incurred in the bonding process is determined by comparison of through section measurements made before and after bonding. The precise width of the nominal 11mm joint is measured and the failure load is thereafter normalised to a standard 11 mm joint width.

In order to conduct an accurate shear stress test it is necessary to avoid bending of the test piece and consequential peel stresses. A special testing rig is used for this purpose. The jig and the test itself have been described in an article at pages 1009-1014 of the Journal of Materials Science 20 (1985). Some test results are presented later in this description.

A variation of the process described above is used to produce an alternative form of lap joint for shear testing. This is described here with reference to Figures 3 and 4 and the test piece is shown in Figure 5. This test piece is designated herein as the "type 2" test piece.

In bonding these test pieces no bonding jig is required for the form of the component pieces is such as to maintain alignment of the pieces. One piece (designated 11 in Figure 3) is placed directly on the lower press platten 4 and the other piece 12 is supported over it by means of supports 13 comprising folded aluminium foil, such that there is a space between the two pieces 11 and 12. There is a machined step 14 which is 15 mm long, extending over the full width of piece 11. This serves to provide accurate control of area and deformation during bonding. The step height is between 0.3 and 0.5mm according to the required degree of deformation. The surface of the step 14 and the whole lower surface of the upper piece 12 are subjected to the same pretreatment described above. The loading/temperature schedule is also the same as that detailed previously. The bonded blank is depicted and dimensioned in Figure 4.

To produce the type 2 test pieces from the bonded blank the latter is cut to size and drilled to provide through-holes 20 for

attachment to the testing apparatus. Slots 21 are cut across the width of the test piece from the surface to the bond line so that the two ends of the test piece remain joined by a joint length " $\ell$ " which is delimited by the slots and hence accurately defined.

5 The slotted test pieces are side trimmed to give the same plan form as the type 1 test pieces and are subjected to the same solution treatment and ageing also. The type 2 test pieces are tested in the same jig as used for the type 1 test pieces. The tables below gives  
10 shear strength test results for type 1 and type 2 test pieces collated by sheet thickness. Save where indicated all samples have been diamond polished rather than silicon carbide polished. All data for 2.5 mm sheet are for test pieces having a loading axis aligned to the rolling direction of the sheet. The 4mm sheet data is all for type 2  
15 test pieces and the designation "L" for Longitudinal or "T" for transverse indicates that the test piece loading axis is respectively parallel with or transverse to the sheet rolling axis.

SHEAR STRENGTH DATA FOR JOINED PIECES OF  
2.5 mm GAUGE 8090 SHEET

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Test Piece Type	Joint Overlap $\ell$ (mm)	Bonding time at 560°C, (h)	Deformation (%)	Shear stress at failure (MPa)	Failure load normalised to 11mm width, (kN)
1	4.4	4	5.3	118	5.7
1*	4.7	1	3.9	119	6.1
1	5.0	1		126	7.1
2	5.1	4	4.8	99	6.4
2	5.2	19	6.1	110	6.3

\* - SiC 1200 grade grit polished

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**SHEAR STRENGTH DATA FOR JOINED PIECES OF  
4 mm GAUGE 8090 SHEET**

5	Test Piece Direction	Overlap $\ell$	Shear stress at failure	Failure load normalised to 11mm width,
		(mm)	(MPa)	(kN)
10	L	1.9	184	3.9
	L	2	202	4.5
	T	2	193	4.3
	T	2	181	4.0
	T	2.1	181	4.1
15	T	2.1	197	4.5
	L	3.0	199	4.7
	L	3.0	177	5.8
	L	3.1	184	6.1
	L	4.9	116	5.0
20	L	4.9	119	6.5
	L	15.1	67	11.1

25 All the above 4mm sheet data is for test pieces having a bonding time at 560°C of 4 hours. The span of bonding deformation is 8-12% of original thickness.

30 To obtain an indication of the bond strength in shear in comparison with parent metal strength in the same loading condition and after the same heating cycle etc, one piece blanks to the type 2 test piece configuration were made and subjected to the same thermal cycling (with 4 hours at 560°C) and recovery heat treatment. Failure loads (normalised to 11mm joint width) spanned 4.5 to 5.4 KN and shear stresses spanned 160-209 MPa (both according to joint overlap). It is clear from this that the bonded joints produce a high proportion of parent metal strengths in shear and this is a good indication of the  
35 suitability of the joints for normal structural applications.

In the DBSPF process pieces of metal are first diffusion bonded to one another over restricted areas then blown apart using argon at superplastic strain rates in the unbonded areas between the diffusion bonds. The superplastic forming is performed with the bonded materials in a hot condition and is such as to subject the bonds to peel stresses rather than shear stress although the finished products might be expected to require shear strength of the bonds. Typically the superplastic forming regime for 8090 alloy spans the 515 to 530°C temperature range and strain rates is around  $3$  to  $6 \times 10^{-4}/S$ .

Peel stress test pieces have been bonded by the diffusion bonding process as described above and tested under various conditions including tests at temperature in the as-bonded condition which is representative of the conditions imposed by DBSPF. All test pieces were pre-polished by diamond powder rather than silicon carbide grit and all were held at 560°C under a predetermined pressure in the band 0.75-1.5 MPa pressure for four hours. The test pieces each comprise two juxtaposed strips bonded together for part of their length. These are reheated to 530°C after bonding to permit the unbonded ends to be pulled apart to form a "T" peel test piece. Some of these were reheated-treated to a solution treated or a solution treated and aged condition. Results for peel tests at various strain rates, test temperatures and metallurgical conditions are presented below.

#### PEEL TEST IN AS-BONDED CONDITION

Test temperature	Strain rate	Peel strength (N/mm)
22°C		50-70
515°C	$\sim 3 \times 10^{-4}/S$	4.4-5
530°C	$\sim 3 \times 10^{-4}/S$	5.2-5.4
515°C	$\sim 3 \times 10^{-3}/S$	9.1
515°C	$\sim 3 \times 10^{-5}/S$	2.7
530°C	$\sim 3 \times 10^{-3}/S$	9.5
530°C	$\sim 3 \times 10^{-5}/S$	2.7

(20 minutes at 530°C then water quenched) gave a peel strength of 70 N/mm. A room temperature peel test on solution treated and aged specimens (aged for 5 hours at 185°C after solution heat treatment as above) gave peel strengths in the range 31-48 N/mm.

5 From the above results it is apparent that the peel strength varies considerably with the metallurgical condition of the material (aged/as bonded) and that it is sensitive to strain rate for elevated temperature tests in the as-bonded condition. Nevertheless the peel strengths obtained are sufficient to permit superplastic forming  
10 without joint peel when the formed section is sufficiently thin, say 1 mm or less.

The invention has also been applied in joining pieces of 8090 alloy reinforced with 20% by weight of SiC grit. All joints were made to the type 2 testpiece configuration, and were made using the same  
15 bonding time/temperature/pressure schedule as described previously. All joints were subjected to a resolutioning for 20 minutes at 530°C followed by ageing for 16 hours at 150°C. Some joints, as identified below, were made without any foil interlayer. One joint was made using a 8090 alloy interlayer and one other joint was made using a  
20 copper foil interlayer. One joint was made for comparison purposes using the same procedure as claimed herein in conjunction with a pre joining plating with copper.

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## SHEAR STRESS TESTS ON BONDED REINFORCED COMPOSITES

	Joint type	overlap length $l$ (mm)	failure shear stress (MPa)	failure mode
5	(a) Composite to composite joints tested at 20°C			
	without interlayer	2.1	116	bond interface
10	"	2.4	86	" "
	"	4.9	>83	base metal
15	50 $\mu$ foil interlayer of 8090 alloy	2.1	149	bond interface
	8 $\mu$ m foil interlayer of copper	2.1	>167	base metal
20	8 $\mu$ m foil interlayer of copper with 1 $\mu$ m copper undercoating	2.3	>142	base metal
25	(b) Composite to composite joints tested at 250°C			
	without interlayer	2.3	150	bond interface
30	(c) Composite to non-composite joint tested at 20°C			
	without interlayer	2.2	138	bond interface

These examples indicate that it is quite possible to secure satisfactory bond strengths with reinforced composites. The bond strength is increased when an interlayer is used as might be expected.



We have found also that those bonds between unreinforced alloys which have been subjected to peel testing in a regime which has caused flow rather than rupture have lost their planar bond line as a consequence of that flow. This suggests that the bonding method might be improved still further by working them at suitable temperatures eg by stretching the materials by 10-20% along the plane of the bond line at temperatures of 100°C plus. Selection of an appropriate temperature will depend upon whether a recrystallised zone can be tolerated. An extensive recrystallised zone would be prejudicial to subsequent superplastic forming. To avoid recrystallisation a working temperature in the region 200-250°C is suggested. In the general case on upper limit for the working temperature would be 500°C or thereabouts - just below the solution treatment temperature of the material.

## CLAIMS

1. A process by which two or more aluminium-lithium alloy pieces are bonded together by the following steps in sequence.

machining or abrading material from each faying surface to a depth sufficient to remove existing surface oxide and the sub-surface zone of lithium-depleted material where such exists;

grinding or polishing the faying surfaces to provide a smoothed surfaces for mating and cleaning these surfaces of polishing residues;

locating the pieces in a vacuum chamber and evacuating the chamber whilst the faying surfaces are either kept apart or under light contact pressure to allow desorption of surface contaminants;

pressing the pieces together heavily at the faying surfaces whilst maintaining the vacuum and increasing the temperature of the pieces in a regime in which the pressure is eased as the temperature is raised; and;

holding the pieces together under light pressure for at least one hour at a temperature in the range of 500 to 580°C.

2. A process as claimed in claim 1 in which a foil interlayer of an aluminium-lithium alloy is placed between the faying surfaces prior to locating the pieces in the vacuum chamber.

3. A process as claimed in claim 1 in which a foil interlayer of copper or silver is placed between the faying surfaces prior to locating the pieces in the vacuum chamber.

4. A process as claimed in any one of the preceding claims in which the pieces are pressed together after evacuation of the vacuum chamber at an initial heavy pressure of 100-120 MPa.

5. A process as claimed in any one of the preceding claims in which the pieces are pressed together in the light pressure phase at a pressure of 0.5 to 3 MPa.
6. A process as claimed in any one of the preceding claims incorporating the additional step, imposed after completion of the others, of stretching the bonded pieces along the plane of the bond line by 10 to 20% of original length at a temperature of 100-500°C.